

Disorder-induced Purcell enhancement in nanoparticle chains

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In this paper we report on numerical study of plasmonic nanoparticle chains with long-range dipole-dipole interaction. We have shown that introduction of positional disorder gives a peak in the density of resonant states (DOS) at the frequency of individual nanoparticle resonance. This peak is referred to Dyson singularity in one-dimensional disordered structures [Dyson F., 1953] and, according to our calculations, governs the spectral properties of local DOS. This provides disorder-induced Purcell enhancement that can find its applications in random lasers and for SERS spectroscopy. We stress that this effect relates not only to plasmonic nanoparticles but to an arbitrary chain of nanoparticles or atoms with resonant polarizabilities.

I. INTRODUCTION

Plasmonic and dielectric nanoparticle chains have been actively studied due to their subwavelength waveguiding properties in a number of papers for the last decade [1–6]. Rapidly developing self-assembly fabrication methods allow production of tens nanometers scale structures with simple and cost-effective techniques [7–11]. One of the main features of self-assembly methods is the randomness and disorder in fabricated structures. Spatial order and periodicity play key role in the process of efficient energy transport, and introduction of disorder leads to suppressing the transmission efficiency [12–14]. However, the role of disorder in photonics and plasmonics have been recently reconsidered. The experimental advances in random lasing [15, 16] stimulated studies on light transport in disorder media [17] and photon management in strongly scattering media [18, 19]. In this paper we discuss the utilization of disorder to induce Purcell enhancement in resonant chains.

Since the early works of Mott and Anderson one-dimensional (1D) disordered structures have been attracting intensive interest of the researchers. Worth noting is the monography of Gredeskul, Pastur, and Pitaevski [20] almost fully dedicated to disorder in 1D. In photonics disordered one-dimensional photonic crystal have been studied in a number of papers [21–23]. We focus on nanoparticle chains that are quasi one-dimensional as they are embedded in 3D, but the chain excitations propagate along the chain direction only. In this prospective it was shown [12, 13] that introduction of disorder in nanoparticle chains stimulates scattering and increase losses. On the other hand, the randomness in plasmonic structures can be beneficial and give rise into giant fluctuations of local fields and to accumulation of energy in “hot spots” [24, 25] that finds its application in SERS.

In this paper we report on how disorder can be utilized to control and engineer optical properties of reso-

nant nanoparticle chain. We demonstrate that in one-dimensional chain the positional disorder stimulates formation of special modes that gives its contribution to DOS. Such behavior was predicted by F. Dyson in 1953 [26] for a chains of mechanical oscillators with random values of spring stiffness. He showed that at zero energy there exists divergence in DOS function. The divergent DOS is related to phonon spectra of solids [27, 28], excitonic structures in disordered 1D J-aggregates [29–31], and mathematically all these systems can be merged within the theory of random matrices [32, 33]. Being disorder induced and, in this sense, disorder protected, the effect of divergent DOS can be implemented for local density of states (LDOS) control and spontaneous emission engineering via Purcell effect [34–36]. This can open a route for fabrication of random lasers in one-dimensional structures and for additional SERS enhancement on disorder plasmonic chains.

The manuscript is structured as follows: in section I we formulate our approach to the problem considering disordered nanoparticle chain as an array of radiating plasmonic dipoles. In section II the problem is treated in quasi statical (QS) and nearest neighbor (NN) interaction limits. We show that there exists a singularity in the DOS function at the frequency of an isolated nanoparticle resonance. The considered simple model partially explains the basic physics lying beyond the discussed effects. In section III more realistic approach with long-range interaction and retardation effects is proposed. We include near, intermediate, and far fields into consideration and show that the Dyson peculiarity still persists in the DOS spectra. Finally, in the section IV we will discuss the Dyson peak in LDOS and demonstrate how it is influenced by losses. In the Appendix A we describe the method of eigen frequency calculation. In Appendix B The case of non-1D (planar) nanoparticle array is considered on the example of double-line chain. The sufficient difference of DOS spectrum comparing to 1D chain is discussed.

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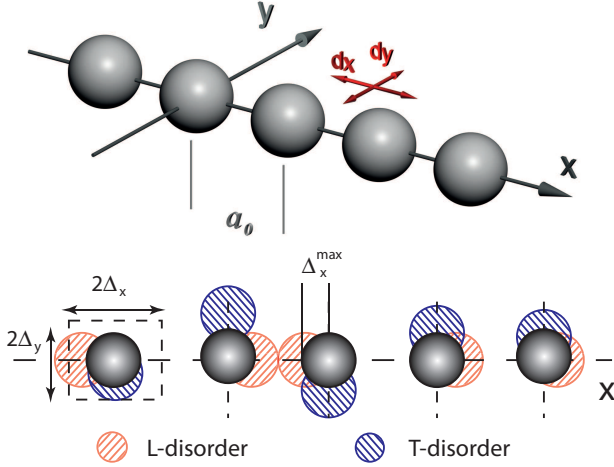


FIG. 1. Regular nanoparticle chain with fixed distance a_0 between nanoparticles (top). The types of the spatial disorder (bottom): longitudinal (L) disorder relates to random position shift in x direction; transverse (T) disorder relates to random position shift in y direction. Δ_x^{\max} is the maximal range of a shift that provides nonoverlapping condition.

II. FORMULATION OF THE PROBLEM

We consider a chain of plasmonic nanoparticles as an example of coupled dipoles with Drude like dielectric permittivity $\varepsilon(\omega) = \varepsilon_\infty - \omega_p^2 / (\omega + i\gamma)$. Here ω_p is the plasma frequency of metal and γ is the damping constant. We consider spherical nanoparticles in vacuum with polarizability

$$\alpha = R^3 \frac{\varepsilon(\omega) - 1}{\varepsilon(\omega) + 2},$$

where R is the nanoparticle radius. Neglecting losses and assuming $\varepsilon_\infty = 1$ the resonant frequency of an individual nanoparticle has the form $\omega_0 = \omega_p / \sqrt{\varepsilon_\infty + 2}$. The polarizability can be written as

$$\alpha(\omega) = R^3 \frac{\omega_0^2}{\omega_0^2 - \omega^2}$$

To account on retardation one can use substitution [37]

$$\frac{1}{\alpha} \rightarrow \frac{1}{\alpha} - \frac{2}{3}ik^3, \quad (1)$$

where k is the wave vector. Dipole interaction of particles splits the resonant frequency ω_0 and takes off the polarization degeneracy. For infinite periodic chain the set of eigen frequencies forms bands for transverse (T) and longitudinal (L) polarization [2, 5, 38]. In order to study the effects of disorder we will consider finite chains consisting of N nanoparticles. We can write down the system of

equations describing the dipole moments of nanoparticle

$$\vec{d}_i = \alpha(\omega) \sum_{j=1, j \neq i}^N \vec{G}_{ij}(\omega) \vec{d}_j, \quad i = 1..N. \quad (2)$$

Here $\vec{G}_{ij}(\omega) = \vec{G}(|\vec{r}_i - \vec{r}_j|, \omega)$ is dyadic Green function [37] that represents the field of a point dipole placed at the coordinate \vec{r}_j and calculated at the point \vec{r}_i . The homogeneous problem describing the system has the form [38–40]

$$\mathbf{H}(\omega) \mathbf{d} = \frac{1}{\alpha(\omega)} \mathbf{d} \quad (3)$$

where \mathbf{H} is $3N \times 3N$ block-matrix, at the ij -th ($1 \leq i, j \leq N$) position there is matrix representation of $\vec{G}(\omega)_{ij}$ tensor, and \mathbf{d} is a block-vector of length $3N$ containing components of \vec{d}_i at i -th position. The homogeneous system (3) has non-trivial solution if the condition

$$\det \left(\mathbf{H}(\omega) - \frac{1}{\alpha(\omega)} \mathbf{I} \right) = 0 \quad (4)$$

is fulfilled, where \mathbf{I} is the unity matrix. Solving this equation one can find all the resonant frequencies of the system [38]. However, for ensembles with large N this appears to be a complicated problem. We propose using perturbation method and represent $\mathbf{H}(\omega) = \mathbf{H}(\omega_0) + \delta \mathbf{H}(\omega_0)$ basing on weak dependence of matrix \mathbf{H} on ω . In this case the inverse polarizability plays the role of $\mathbf{H}(\omega_0)$ matrix eigen number [39, 41]. The eigen frequencies can be found within the first order precision: $\omega_n = \omega_n^0 + \omega_n^1$. More detailed description of this approach is described in the Appendix A.

A. Introduction of disorder

In this paper we consider a plasmonic chain with randomly varying nanoparticle positions that is also referred as non-diagonal disorder. Each nanoparticle can be randomly shifted around its position in the regular chain. We limit the consideration with $x - y$ plane perturbation only, keeping the chain to be planar (see Fig. 1). Such geometry is the most prospective from the point of possible technological realization: nanoparticle ensembles are commonly fabricated on the top of planar substrates [9, 10, 42]. Each nanoparticle center can be varied for a random value dx or dy . We use uniform distribution of the i -th nanoparticle shift $dx_i(dy_i)$ in the range $-\Delta_{x(y)} \leq dx(dy)_i \leq \Delta_{x(y)}$. We introduce the relative parameter of disorder $\delta_{x(y)} = \Delta_{x(y)} / a_0$, which is limited with its maximal value $\delta_{max}(a_0)$ to satisfy the condition of non-overlapping. For instance, the value of $\delta_x^{max} \approx 0.18$ for $a_0 = 3R$.

We define two types of disorder as it is shown in Fig. 1: (a) $\Delta_x \neq 0, \Delta_y = 0$ longitudinal disorder (L-disorder) and (b) $\Delta_x = 0, \Delta_y \neq 0$ transverse disorder (T-disorder).

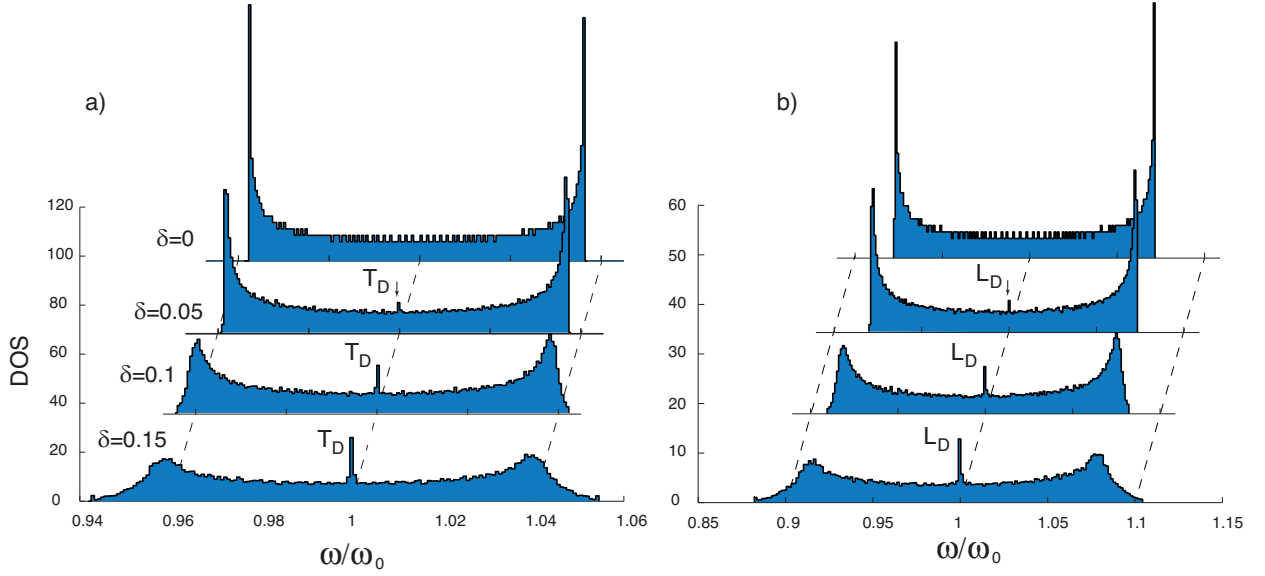


FIG. 2. DOS in the QS limit with amplitude of L-disorder. a) T-polarized modes b) L-polarized modes. $N=800$. The results are calculated for $a_0 = 3R$, $N=800$, and averaged over 50 realizations.

We need to stress that L-disorder keeps splitting of T and L polarizations, meanwhile T-disorder mixes T and L polarizations. We also will discuss the case of TL disorder that is superposition of both types of disorder. We will pay much attention to L-disordered chains as they have simpler description but possess all the key properties of the considered systems.

III. SIMPLE MODEL: QS LIMIT AND NEAREST-NEIGHBOR INTERACTION

We start with the consideration of nanoparticle size and interparticle distance small comparatively to the wavelength (QS limit). The matrix \mathbf{H} depends on frequency through the term $\omega r/c$ that goes to zero in the QS limit. The matrix \mathbf{H} becomes frequency independent and, thus, real symmetric. We come to exact eigen value problem with inverse polarizabilities as eigen values λ_k . The eigen frequency for each eigen value λ_k can be found from the relation $1/\alpha(\omega_k) = \lambda_k$.

$$\mathbf{H}(0)\mathbf{d} = \lambda_k \mathbf{d} = \frac{1}{\alpha_k} \mathbf{d}$$

In the simplest case of NN interaction the matrix \mathbf{H} has the form:

$$\begin{aligned} \mathbf{H}_{i,i} &= 0 \\ \mathbf{H}_{i+1,i} &= \mathbf{H}_{i,i+1} = \frac{-1(2)}{a_{i,i+1}^3} \quad \text{for T(L)-polarization} \end{aligned} \quad (5)$$

For regular chain the distance $a_{i,i+1}$ between the particles is constant $a_{i,i+1} = a_0$ and the dispersion relation

$\omega(q)$ has the form [38]:

$$\begin{aligned} w_{T(L)}^2(q) &= w_0^2 (1 + g_{T(L)} \cos(qa)) , \\ g_T &= 2 \frac{R^3}{a_0^3} \quad g_L = -4 \frac{R^3}{a_0^3} \end{aligned}$$

where q is quasi vector.

The DOS function then can be described by a simple formula that exhibits van Hove singularities at the band edges:

$$DOS_{T(L)}(\omega) = \frac{2\omega}{a_0 \sqrt{\omega_0^4 g_{T(L)}^2 - (\omega^2 - \omega_0^2)^2}}$$

In the case of L-disordered chain $a_{i,i+1} = a_0 + \Delta_{i+1} - \Delta_i$ is a random number with mean value equal to a_0 and Δ_i are the nanoparticle shifts in x direction and are given by uniform probability distribution. Thus, the system is described with two-diagonal symmetric matrix filled with random numbers. Such matrix was considered in the classical paper by F. Dyson [26]. One of the results demonstrated in this paper was a singularity of DOS at the zero eigen values in the limit $N \rightarrow \infty$ for non diagonal disorder that is the case of our study. In terms of polarizability the condition of $\lambda = 0$ is satisfied at the frequency $\omega_D = \omega_0$ at which inverse polarizability has root $1/\alpha(\omega) \sim 1 - \omega^2/\omega_0^2$. In the Fig. 2 the DOS of nanoparticle chain is presented in the QS limit and NN interaction for different value of disorder parameter δ_x . Similar behaviors were already obtained in [29–31] for excitons in one-dimensional disorder structures. The Frenkel hamiltonian in tight binding limit considered in these papers has the form of (5).

The QS nearest-neighbor interaction limit gives the logarithmic divergence of the DOS function in the vicin-

ity of zero eigen value [26]. However, since Dyson classical paper there are various discussions on the nature of the divergent states. In [43] it was shown that the states are extended, and, thus, delocalized. The later studies by [30] dispute this claim. Our calculations of participation ratio also demonstrate that the localization length is finite, however the [31] reports that this may be due to finite size effect. The appearance of this singularity can be understood within the perturbation theory. Kozlov et.al. [30] showed that in QS limit introducing disorder as perturbation gives zero shift of energy levels in the middle of the band that results in increasing of DOS.

Since the early works of Dyson and Wigner active development of random matrix theory has started, and sparse random matrices play an important role among them. In several papers [28, 44, 45] it has been shown that the singularity of DOS for zero eigen value is tightly related to sparsity of random matrices. The constructed matrix (5) is just the case of a sparse random matrix. The random matrix theory predicts that this effect can be observed in the vicinity of zero eigen value in a wide class of random matrices. In our terms, zero value eigen values correspond to resonance in polarizability. Consequently, divergent DOS behavior will be observed in quite general class of interacting resonant oscillators and plasmonic particle chain is just an example.

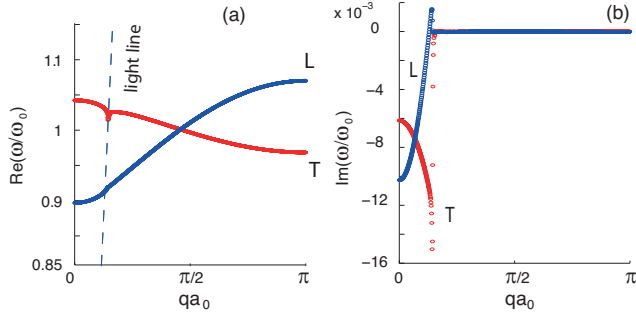


FIG. 3. (a) Dispersion relation of regular nanoparticle chain with retardation for T and L modes. The dispersion of light in free space (light line) is also shown. (b) Dependence of imaginary parts of eigen frequencies on quasi vector for T and L modes. The results are calculated for $a_0 = 3R$, $\gamma = 0$, $\kappa = 0.15$, $N=400$.

IV. RETARDATION AND LONG-RANGE INTERACTION

The previous studies by [29, 30] considered long-range dipole-dipole interaction only in QS regime, i.e. the interaction was governed by near fields that decrease as $\sim 1/r^3$. Considering plasmonic chain the long range radiation effects play important role. We considered fully retarded model with intermediate ($\sim 1/r^2$) and far ($\sim 1/r$) fields that are the major interaction terms at long distances. We demonstrate that despite stronger long-range

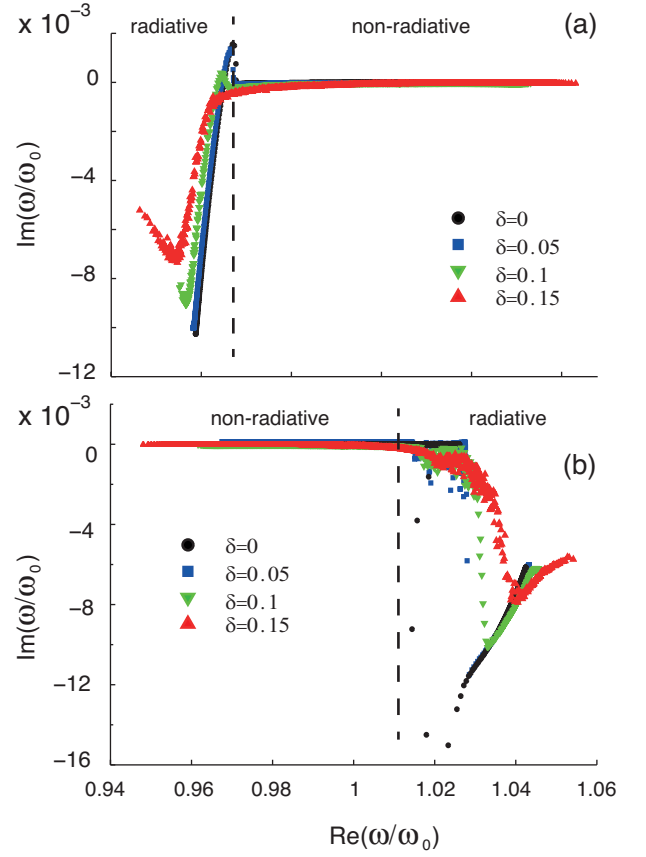


FIG. 4. The imaginary parts of eigen frequencies for (a) L-polarization and (b) T-polarization for different magnitude of L-disorder. The parameters of calculation are taken the same as in the Fig. 3. Results are obtained after averaging over 50 realizations.

coupling one still can observe the peculiarity at the band center.

To include the retardation effects we consider frequency dependent matrix $\mathbf{H}(\omega)$ and use substitution (1) to account on dipole emission that introduces losses in the system. We neglect Drude losses $\gamma = 0$ at this stage, but will add them in the last section of the manuscript. To find the eigen frequency of the system (3) we apply approximate method described in the Appendix A section. The main parameter that defines the dependence of \mathbf{H} matrix on frequency is $\kappa = \omega_0 R/c$. Indeed, the eigen frequencies will lie around ω_0 and parameter κ shows the retardation strength, i.e. typical phase shift at the scales of nanoparticle radius. In this paper we are limited with small values of κ as approximate method of calculation based on perturbation theory diverges for large κ , thus, for all the calculations below we take $\kappa = 0.15$.

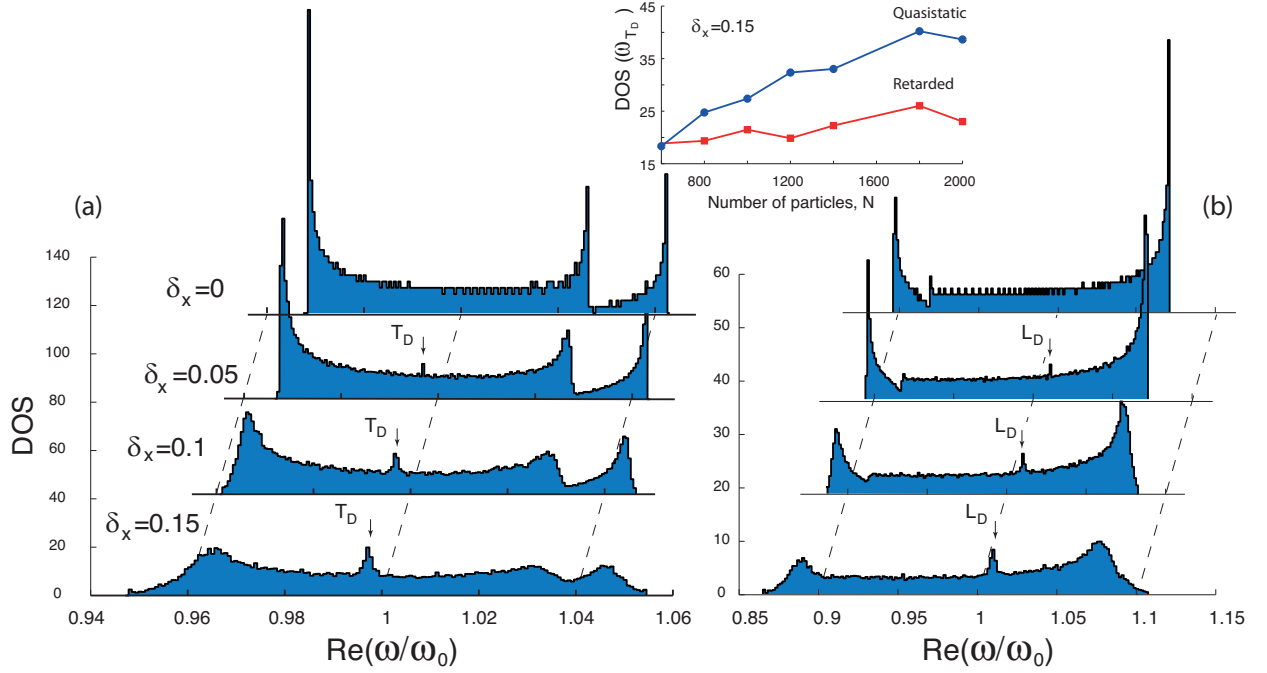


FIG. 5. DOS for L-disordered chain with retardation for T-polarized modes (a) and L-polarized modes (b). The results are calculated for $a_0 = 3R$, $\kappa = 0.15$, $\gamma = 0$, $N = 800$ and averaged over 50 realizations. Inset: DOS calculated at the frequency ω_{T_D} corresponding to T_D peak in quasi state limit (see Fig. 2) (circles) and with retardation and long-range interaction (squares) depending on chain length N at fixed magnitude of disorder $\delta_x = 0.15$.

A. Regular chain

To understand the influence of disorder with account on retardation we again start with the regular chain. The periodical nanoparticle chains with account on retardation and losses have been extensively studied during last decades [5, 13, 38] because of their waveguiding properties and application in plasmonics. The real and imaginary parts of regular chain eigen frequencies are shown in Figure 3 (a,b) for T and L modes. The dispersion relation depicted in Fig. 3 (a) was obtained by sorting the eigenmodes in the descending order of node numbers in the eigen vector. The Bloch vector corresponds to number of zeroes as:

$$q = \frac{\pi(n+1)}{Na_0}$$

According to [38] the T-modes lying above the light line are well coupled to light and are highly radiative (superradiative) that is confirmed by the large negative value of $\text{Im}(\omega_i)$. On the contrary, the modes below the light line are non-radiative (waveguiding regime) and are almost decoupled with light however the imaginary part is nonzero either. The strong hybridization of waveguiding modes with light is seen in the vicinity of the light line crossing with the dispersion curve. The calculation method shows discrepancy for L-modes close to $ka \sim 0.2$ where the imaginary part becomes positive.

To demonstrate how disorder affects the eigen spec-

trum we plotted imaginary parts of eigen frequencies in the Fig.4 for T-modes (a) and for L-modes (b). We see that the disordered structures inherit the properties of ordered systems for both T- and L-modes and we again can divide the spectral region into highly radiating and non-radiating regions. Destruction of periodicity increases losses in non-radiative region due to additional scattering of Bloch waves on defects, but in the radiating region losses are decreased because disorder suppresses the superradiative regime.

To describe the properties of disordered structures we again introduce DOS similar to the QS case. However, in the case of retardation the eigen frequencies are complex, and we will plot the DOS depending on real part of eigen frequency $\text{Re}(\omega)$ (see Fig. 5).

The DOS function for regular chain has complicated structure due to the strong hybridization of chain modes with light. With the increase of disordered the DOS function becomes more and more homogeneous. But despite the long-range interaction and retardation one can see the divergence of DOS function near the frequency ω_0 similarly to the case of NN limit. However, the DOS peak is now shifted for T and L-modes comparatively to the case shown in Fig. 2 due to long-range interaction effects. The shift of the peak corresponds to the shift of band central frequency [30] and this shift has opposite sign for T and L modes. The DOS peak intensity at increases with the increase of chain length N (see Fig. 5 inset) both in QS and retarded case which reflects it

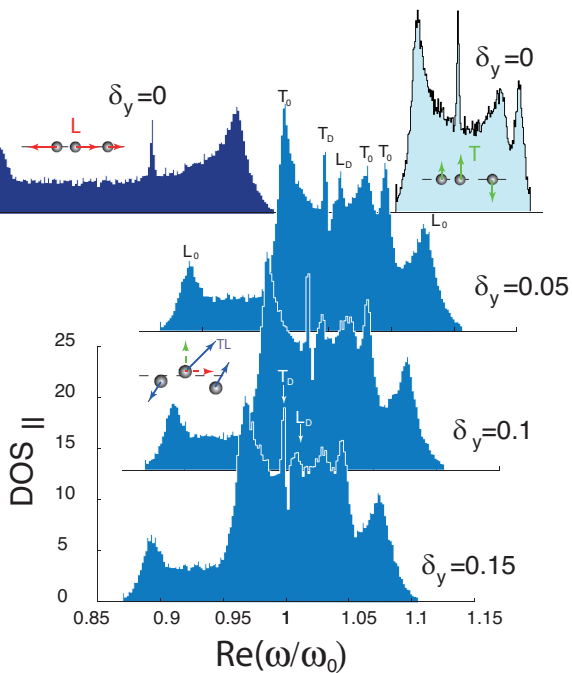


FIG. 6. DOS function for TL-disordered chain with $\delta_x = 0.15$ and different values of δ_y . The DOS functions of T and L polarized states for purely L-disordered ($\delta_y = 0$) chain are shown separately at the top of the figure. Each dipole moment in TL-disordered mode has x and y components (see inset). For weak disorder one can divide modes into mainly T- and mainly L-polarized. The peaks related to DOS of ordered chain are denoted as T_0 and L_0 along with Dyson peaks T_D and L_D . The parameters of calculations are the same as in the Fig. 5.

divergent character.

V. PLANAR (TL) DISORDER

Till now we have considered only L-disordered that conserves splitting of modes into T and L polarizations. Introduction of TL-disorder partially mixes mode polarization. The polarization of eigen modes lying in the x - y plane will be mixed, and the z -component of vector will be still independent and behave similarly to y -polarized modes in the case of L-disorder. Thus, we will not discuss z -polarized modes in this section. To emphasize that we will refer to $\text{DOS}_{||}$ meaning the density of resonant states with polarization vector lying in the x - y plane. The influence of TL-disorder on DOS is depicted in the Fig. 6. One can see that introduction of weak disorder $\delta_y = 0.05$ mixes the polarization not significantly and the DOS function is roughly the sum of T (light blue) and L-polarization (dark blue) DOS functions in purely L-disordered chain. We mark the peaks originating from the ordered T and L polarizations as T_0 and L_0 respectively. The Dyson peaks are marked as T_D and L_D . One

can see that the increase of T-disorder affects the L_D peak mainly and for $\delta_y = 0.15$ the L_D is almost smeared out. The T_D peak on the contrary does not change significantly. Here we can conclude that the transversal Dyson mode (T_D) is stable with respect to transversal disorder in contrary with longitudinal mode.

In the Appendix B we have also considered the special case of double-line chain with TL disorder to demonstrate that the features of Dyson singularity originates from 1D character of interaction. Considering two parallel chains of nanoparticle dramatically changes the DOS function and the fine structure around ω_0 disappears.

VI. LOCAL DOS AND PURCELL ENHANCEMENT

We have demonstrated that the disorder in nanoparticle chain leads to peculiarity in general DOS function near the band center, which relates to Dyson singularity of 1D disordered system. In nanophotonics the role of LDOS function is even of more importance. The LDOS is connected to DOS via simple relation $D(\omega) = \int \rho(\mathbf{r}, \omega) d^3\mathbf{r}$, thus, one can expect that LDOS properties possess similar spectral features as DOS.

To obtain the LDOS function we have calculated the dyadic Green function $\hat{G}(\mathbf{r}, \mathbf{r}, \omega)$ of disordered chain [37]. We have placed the electric dipole source at the position above the chain, as it is shown in Fig.7, at the center of periodic chain at the coordinate $(0, 0, 2R)$, and calculated chain response to the excitation by the source in dipole approximation. The x , y , and z components of electric field at the point of x , y , and z -oriented dipole source correspondingly give us the diagonal elements of the dyadic tensor, and the LDOS can be found as:

$$\rho(\mathbf{r}, \omega) = \rho_0 + \frac{2\omega}{\pi c^2} \text{Im} \left(\text{Tr} \hat{G}(\mathbf{r}, \mathbf{r}, \omega) \right)$$

We normalize LDOS over $\rho_0 = \omega^2 / \pi^2 c^3$ that is LDOS in vacuum that gives us the Purcell enhancement (PE) factor [34] averaged over dipole orientations. The calculated LDOS is depicted in Fig.7 for TL disordered chain. The spectrum lines are plotted for different values of losses in metal. For $\gamma = 0$ only retardation losses are accounted. Low losses lead to strong fluctuations of the PE due to very narrow resonances. The fluctuations are particularly strong for the frequency range below the ω_0 that corresponds to non-radiative regime of T-modes for periodic chain where radiative losses are small (see Fig.3 (a)). Adding Drude losses we significantly suppress the fluctuations and achieve smooth LDOS line already for several hundreds of iterations. One can see that the spectrum of PE replicates the DOS spectrum of T-modes (see fig.5 (b)) with clear Dyson peak. It is relatively weak as the length of the chain was taken $N = 101$ to reduce computation time (DOS function presented in Fig. 5 was computed for $N = 800$). Absence of L_D peak relates to weak coupling of dipole source with L-modes. Indeed,

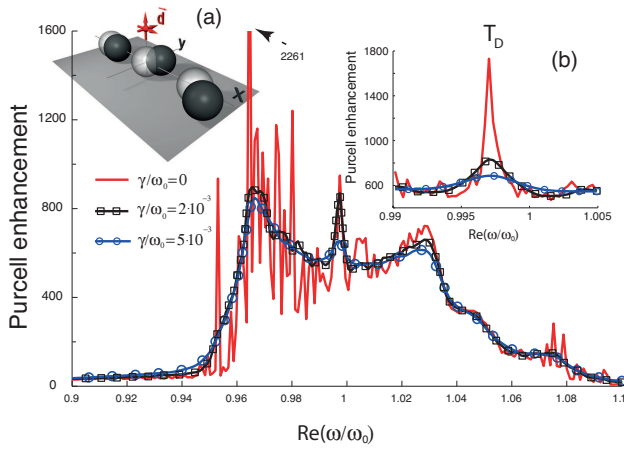


FIG. 7. Spectrum of Purcell enhancement factor calculated at the point $(0, 0, 2R)$ and plotted for different values of losses in metal. Radiation losses are also accounted. *Inset*: a) the point of Purcell enhancement calculation; b) zoomed Purcell enhancement curve around Dyson singularity. The number of particles in chain $N = 101$. The parameters of calculation: $\delta_x = 0.15$, $\delta_y = 0.15$, $\kappa = 0.15$. The $\gamma = 0$ curve was obtained after averaging over 2000 realizations, and the other curves were averaged over 500 realizations.

coupling to L modes occurs only via near fields only for a dipole polarized in x direction. The coupling via far field is suppressed as there are no x -components of electric field.

The spatial distribution of PE depending on dipole source position is plotted in the Fig. 8 for $z > R$ at the frequency corresponding to T_D . Recalling that LDOS should be periodic in x direction, we plot PE distribution over one unit cell only. The calculated PE is symmetric in the unit cell in the $x-y$ plane respectively to nanoparticle position in regular chain. Thus, we plot only half of the unit cell cross-section in $x-z$ and $y-z$ planes as shown in Fig. 8 inset. One can see that the PE distribution rapidly decrease away from the chain. We need to mention that the dipole model should not be valid close to nanoparticle surface around $z \lesssim R$ and $x \approx 0$, $y \approx 0$.

We again would like to stress that the calculated LDOS corresponds to long range-interacted and retarded system. The presence of peculiarity close to nanoparticle resonance ω_0 is related to existence of resonant behavior of polarizability (existence of a root in the inverse polarizability $1/\alpha$). Thus, we predict the similar LDOS properties for a general class of resonantly interacting oscillatory chain of different nature.

VII. CONCLUSION

We have studied resonant nanoparticle chain with positional (non-diagonal) disorder. We have shown that disorder induces the divergence in DOS function in the

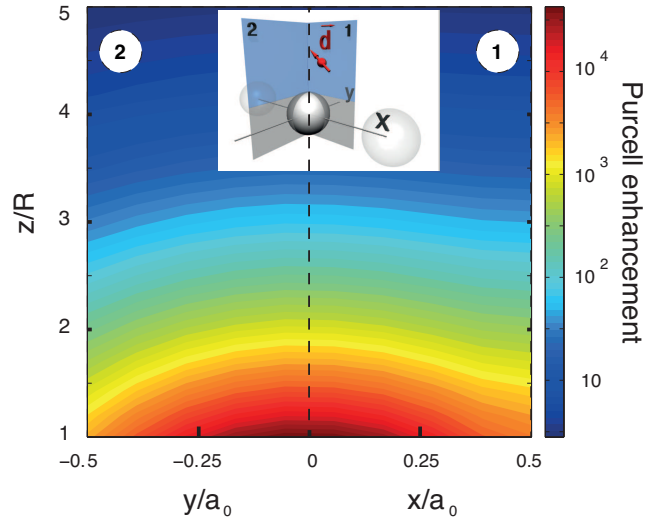


FIG. 8. LDOS enhancement distribution for TL disordered chain at the frequency of Dyson singularity pointed by arrow in the fig. 8. Due to the symmetry of the problem LDOS is plotted only in a half of the unit cell as shown in the *inset* (planes 1 and 2) for $z \geq R$. The number of particles in chain $N = 101$. The parameters of calculation: $\delta_x = 0.15$, $\delta_y = 0.15$, $\kappa = 0.15$, $\gamma/\omega_0 = 2 \cdot 10^{-3}$. The result was obtained after averaging over 500 realizations.

vicinity of individual nanoparticle resonance that is associated with Dyson singularity. This divergence is stronger in the quasi statical, nearest neighbor interaction limit, but also was found in long-range interacting system with retardation effects. Such DOS structure is an internal feature of one-dimensional systems and can not be observed in 2D and 3D arrays. We have shown that positional disorder affects T and L modes differently that is related to the character of dipole-dipole coupling. The LDOS function inherits the structure of DOS and has a peak in the vicinity of an individual nanoparticle resonance. This results may be applied for the observation of Purcell enhancement and for the development of spontaneous time emission engineering in general case of interacting oscillators with resonant polarizabilities. We believe that this effect can be applied for random lasing in one-dimensional structures and for additional enhancement of SERS signal in the vicinity of disordered chains.

VIII. ACKNOWLEDGEMENT

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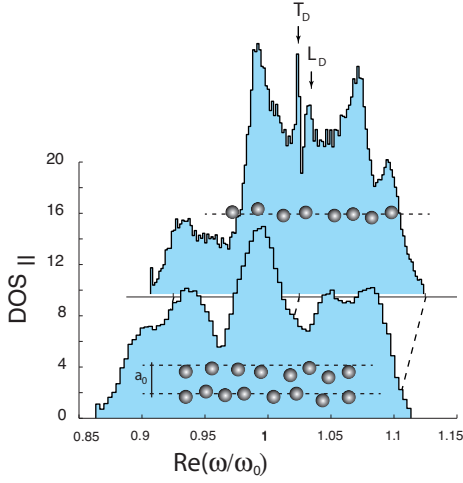


FIG. 9. Comparison of $DOS_{||}$ functions for double nanoparticle chain with $N = 400$ and nanoparticle array $2 \times N$ calculated in QS limit. The disorder parameters $\delta_x = 0.15$ $\delta_y = 0.15$. The separation distance between the chain lines is a_0 .

IX. APPENDIX

Appendix A: Calculation method

To solve the system (3) we imply weak dependence of the matrix \mathbf{H} on frequency ω and decompose it into a series around the frequency $\omega = \omega_0$:

$$\mathbf{H}(\omega) = \mathbf{H}(\omega_0) + \delta\mathbf{H}(\omega_0) \cdot (\omega - \omega_0) + \dots = \mathbf{H}_0 + \delta\mathbf{H}_0 \cdot (\omega - \omega_0) + \dots$$

The matrix \mathbf{H}_0 is matrix $\mathbf{H}(\omega)$ calculated at the frequency $\omega = \omega_0$. The zero order eigen values can be

computed from the system

$$\mathbf{H}_0 \mathbf{d} = \lambda^{(0)} d. \quad (\text{A1})$$

The inverse polarizability is a zero order eigen value and the set of eigen frequencies are computed from relation $1/\alpha(\omega_i^{(0)}) = \lambda_i^{(0)}$, where i is the number of eigen value and index (0) denotes zero-order approximation. Then we assume that $\lambda_i \approx \lambda_i^{(0)} + \lambda_i^{(1)}$ and the first order corrections $\lambda_i^{(1)}$ is computed with perturbation method

$$\lambda_i^{(1)} = \langle \bar{d}_i^0 | \delta\mathbf{H}_0 \cdot (\omega_i - \omega_0) | d_i^0 \rangle.$$

Here we use quasi-scalar product $\langle \bar{a} | b \rangle = \sum_i a_i b_i$ as matrix \mathbf{H} is complex symmetric but non-Hermitian [41].

Such approach is valid as far as the perturbation term $\delta\mathbf{H}_0 \cdot (\omega - \omega_0)$ is small. The matrix $\mathbf{H}(\omega)$ depends on frequency in the form of kr , and $\delta\mathbf{H}(\omega) \sim R/c\mathbf{H}(\omega)$. The perturbation term is smaller by factor of $\omega_0 R/c(\omega_i - \omega_0)/\omega_0 = \kappa\delta\omega_i$ than \mathbf{H}_0 . From QS limit the estimation of band width gives $\delta\omega \sim (R/a_0)^3$. Thus, the perturbation method should be valid if $\kappa(R/a_0)^3 \ll 1$. For the parameters of calculation used in this study $\kappa = 0.15$ and $a_0 = 3R$ this condition is fulfilled.

Appendix B: Two line chain

The dimensionality of the structure plays an important role for the observed DOS peak. To demonstrate that we have calculated the in-plane DOS function (see Section V) for a two line chain of nanoparticles separated with distance a_0 from each other in y -direction. The calculated DOS for in-plane modes in QS limit is shown in fig. (9). One can see that the fine structure of the DOS is totally smeared out for two line chain. Introduction of retardation only enhances this effect. Thus, despite the fact that the system is embedded in 3D the quasi one-dimensional feature of nanoparticle chain is crucial for the observed Dyson peak.

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